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## Linear Absorption and Scattering of Laser Beams

F. X. KNEIZYS  
S. A. CLOUGH  
E. P. SHETTLE

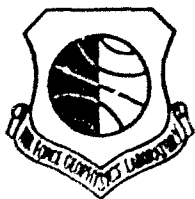
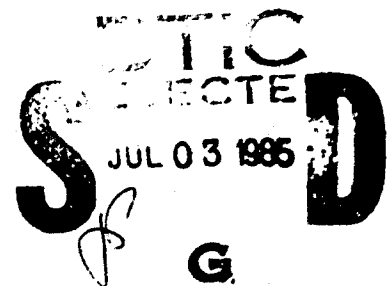
L. S. ROTHMAN  
R. W. FENN



27 September 1984



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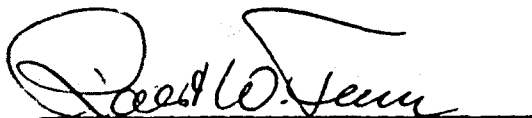
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<p>This report describes the linear transmission effects of the atmosphere on laser beams and gives examples of high resolution (FASCODE) model predictions of transmittances for CO<sub>2</sub>, DF, HF, Nd-Yag, Iodine, and Excimer laser wavelengths. Keywords:</p> <p>Neodymium - Yttrium Aluminum Garnet Hydrogen Fluoride Deuterium Fluoride Carbon dioxide</p>				
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# Linear Absorption and Scattering of Laser Beams

## 1. INTRODUCTION

The attenuation of laser radiation in the earth's atmosphere is caused by the absorption and scattering by molecules and aerosols along the atmospheric path. Molecular line and continuum absorption depend on the amount of absorbing gas, the temperature, and the density of the line broadening gases along the path. The principal molecular absorbers are water vapor, carbon dioxide, ozone, nitrous oxide, carbon monoxide, and methane. Molecular nitrogen and oxygen are less important absorbers, even though they are the principal constituents. Molecular line absorption is a complex and highly variable function of wavenumber\* due to the numerous rotation and vibration-rotation transitions and transition line shapes of the atmospheric molecules. In addition to molecular absorption, one must consider molecular scattering, aerosol absorption, and aerosol scattering. These have a much weaker spectral dependence than molecular absorption. Molecular (Rayleigh) scattering depends on the density and refractive index of the gases along the path. Aerosol absorption and scattering are functions of the number density and size distribution of the particles as well as their complex refractive index and to a lesser extent their shape.

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\*The wavenumber  $\nu$  (in  $\text{cm}^{-1}$ ) is the unit of measurement used in spectroscopy. It is the reciprocal of wavelength.

Although these absorption and scattering processes are linear, they can play a role in nonlinear phenomena. Thermal blooming is caused by absorption of radiation. Dielectric breakdown of air can occur in high energy laser beams in the presence of aerosols.

## 2. MOLECULAR ABSORPTION

The molecular line absorption coefficient  $k_{aML}(\nu)$  at the wavenumber value  $\nu$  ( $\text{cm}^{-1}$ ), assuming superposition of individual spectral transitions, is given by

$$k_{aML}(\nu) = \nu \tanh(hc\nu/2kT) \sum_i \rho(m_i) S_i(T) [f(\nu, \nu_i) + f(\nu, -\nu_i)] \quad (1)$$

where  $\rho(m_i)$  ( $\text{mol}/\text{cm}^3$ ) is the number density of the molecular species  $m_i$ , with transition wavenumber  $\nu_i$ , and  $S_i(T)$  [ $1/(\text{mol}/\text{cm}^2)$ ] is the intensity at temperature  $T$  ( $^{\circ}\text{K}$ ) appropriate to the line shape  $f(\nu, \nu_i)$  ( $1/\text{cm}^{-1}$ ). The quantity,  $\nu \tanh(hc\nu/2kT)$ , is the radiation field dependent term. The formulation in Eq. (1) is an extension of that used by Van Vleck and Huber<sup>1</sup> and discussed by Clough et al.<sup>2</sup>

The intensity  $S_i$ , is expressed in terms of the transition strength  $|\mu^2|_i$  (debye<sup>2</sup>)\* as

$$S_i(T) = \frac{8\pi^3 \times 10^{-36}}{3hc} |\mu^2|_i (1 + e^{-hc\nu_i/kT}) \frac{e^{-hcE_i/kT}}{Q_v(T) Q_R(T)} \quad (2)$$

where  $E_i$  ( $\text{cm}^{-1}$ ) is the lower state energy for the transition and  $Q_v(T)$  and  $Q_R(T)$  are the vibrational and rotational partition functions for the appropriate molecular species.

The line shape function satisfies the normalized condition,

$$\int_{-\infty}^{+\infty} d\nu f(\nu, \nu_i) = 1. \quad (3)$$

1. Van Vleck, J. H., and Huber, D. L. (1977) Absorption, emission and linebreadths: a semihistorical perspective, *Rev. Mod. Phys.* 49:939.
2. Clough, S. A., Kneizys, F. X., Davies, R., Gamache, R., and Tipping, R. (1980) Theoretical line shape for  $\text{H}_2\text{O}$  vapor; application to the continuum; in *Atmospheric Water Vapor* edited by A. Deepak, T. D. Wilkerson and L. H. Ruhnke, Academic Press, New York.

\* 1 debye =  $10^{-18}$  esu cm



The expression for the absorption coefficient, Eq. (1), satisfies two important conditions: the preservation of transition strengths (Nyquist condition) and the condition of radiation balance between emission and absorption for a system in thermal equilibrium.

The line shape function  $f(\nu, \nu_i)$  is dependent on molecular species, broadening density, and temperature. For atmospheric slant paths, the shape changes from a collision-broadened line shape at the surface to a velocity-broadened line shape at the top of the atmosphere. In the intermediate range, a Voigt line shape must be used.

The collision-broadened line shape is given by

$$f_C(\nu, \nu_i, \alpha_i^C) = \frac{1}{\pi} \frac{\alpha_i^C}{(\nu - \nu_i)^2 + (\alpha_i^C)^2} \quad (4)$$

where  $\alpha_i^C$  ( $\text{cm}^{-1}$ ) is the collision-broadened halfwidth at half maximum. The collision-broadened halfwidth depends on the number density,  $\rho$ , and temperature,  $T$ , of the absorbing medium and is given by the relation

$$\alpha_i^C(\rho, T) = \alpha_i^C(\rho_0, T_0) (T/T_0)^x (\rho/\rho_0) \quad (5)$$

where  $\alpha_i^C(\rho_0, T_0)$  is the collision halfwidth at a reference number density  $\rho_0$  and temperature  $T_0$  and the quantity  $x$  characterizes the temperature dependence of the halfwidth. Typical values of  $x$  range between 0 and 0.5 depending on molecular species, with 0.5 representing the classical temperature dependence of the thermal velocity.

It is now established that the impact line shape given by Eq. (4) is appropriate only to the central portion of the spectral lines and that the line shapes in the wings deviate significantly from the impact result. It is known rigorously that the far wings of collision broadened line shapes must decrease exponentially. Deviations from collision-broadened line shapes are modeled through empirical functions and these effects are included in the molecular continuum absorption coefficient described below in Eq. (10) for water vapor and carbon dioxide.

For the Doppler regime, where velocity broadening dominates, the line shape function is

$$f_D(\nu, \nu_i, \alpha_i^D) = \frac{1}{\alpha_i^D} \left( \frac{\ln 2}{\pi} \right)^{\frac{1}{2}} \exp [-\ln 2 (\nu - \nu_i / \alpha_i^D)^2] \quad (6)$$

and the Doppler halfwidth at half maximum is defined as

$$\alpha_i^D = \frac{\nu_i}{c} \left[ 2 (\ln 2) \frac{kT}{M/N_0} \right]^{1/2} \quad (7)$$

where  $M$  is the gram molecular weight of the molecular species and  $N_0$  is Avogadro's number.

For the intermediate regime, the Voigt line shape is given by the convolution of the collisional and velocity broadened functions and can be written as

$$f_V(\nu, \nu_i, \alpha_i^C, \alpha_i^D) = \int_{-\infty}^{\infty} d\nu' f_D(\nu', \nu_i, \alpha_i^D) f_C(\nu - \nu', \nu_i, \alpha_i^C) \quad (8)$$

assuming the two broadening processes are independent.

A Voigt parameter,  $\zeta$ , is defined in terms of the collisional and Doppler half-widths,

$$\zeta = \frac{\alpha_i^C}{\alpha_i^C + \alpha_i^D} \quad (9)$$

for which  $\zeta = 0$  in the Doppler limit and  $\zeta = 1$  in the collision broadened limit.

Figure 1 shows the Voigt parameter,  $\zeta$ , as function of altitude and wavenumber for the U.S. Standard Atmosphere.

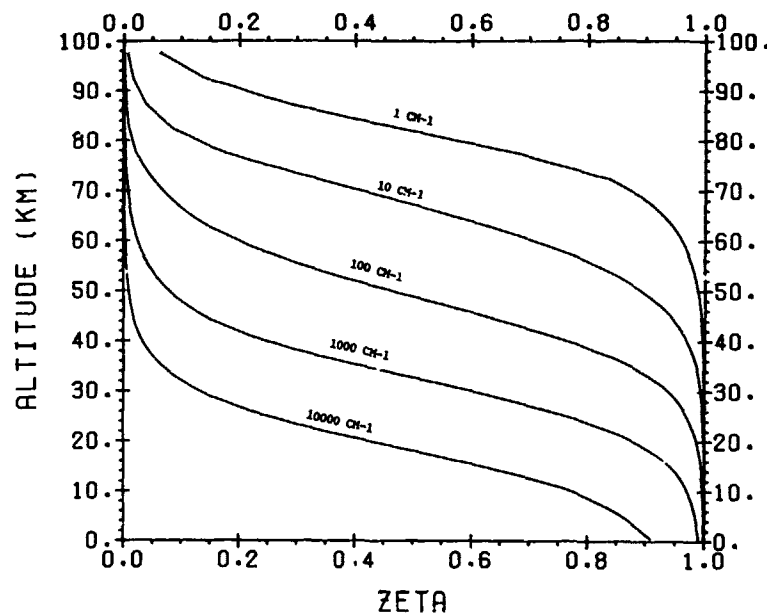


Figure 1. The Voigt Parameter Zeta, as a Function of Altitude and Wavenumber for the U.S. Standard Atmosphere

In addition to line absorption, molecular continuum absorption, such as that due to water vapor, carbon dioxide, and collision-induced absorption by nitrogen, must be included in the attenuation calculation. The molecular continuum contribution to the absorption coefficient,  $k_{aMC}(\nu)$  is given by the expression

$$k_{aMC}(\nu) = \rho_s \nu \tanh(hc\nu/2kT) [(\rho_s/\rho_o) C_s(\nu, T) + (\rho_f/\rho_o) C_f(\nu, T)] \quad (10)$$

where  $\rho_s$  is the density of the absorbing molecular species,  $(\rho_s/\rho_o)$  and  $(\rho_f/\rho_o)$  are the number density ratios for the self and foreign continua,  $\rho_o$  is a reference number density, and  $C_s$  and  $C_f$  [ $(\text{cm}^{-1} \text{ mol}/\text{cm}^2)^{-1}$ ] are wavenumber dependent continuum absorption parameters for the self and foreign components.

The total molecular absorption coefficient  $k_{aM}(\nu)$ , is the sum of the line and continuum contributions [Eqs. (1) and (10)] and is defined as

$$k_{aM}(\nu) = k_{aML}(\nu) + k_{aMC}(\nu). \quad (11)$$

High resolution laser transmission calculations require detailed knowledge of the characteristics of the individual absorption lines of atmospheric gases: line position, line intensity, line width, and shape. The AFGL Line Parameters Compilation<sup>3</sup> contains the related parameters for the major atmospheric absorbing gases: water vapor, carbon dioxide, ozone, nitrous oxide, carbon monoxide, methane, and oxygen. Another compilation covers a variety of trace gases present in the atmosphere.<sup>4</sup> These line compilations cover the spectral range from 0.5  $\mu\text{m}$  through the mm-wave region, and together represent a third of a million transitions.

The species presently covered in these data bases are summarized in Table 1. The isotopes are abbreviated by the code 161  $\equiv \text{H}_2^{16}\text{O}$ , 162  $\equiv \text{HD}^{16}\text{O}$ , and so on. The first seven species (incorporated in the Main Atlas) include all transitions contributing to at least 10 percent absorption over a maximum atmospheric path. The standard temperature chosen is 296 K. The gases in the Trace Gas Compilation are of importance to a variety of problems including stratospheric probing, pollution monitoring, temperature profile retrieval, and laboratory studies. The Main Atlas covers the frequency range 0 to 20,000  $\text{cm}^{-1}$ , the low frequency domain

3. Rothman, L. S., Gamache, R. R., Barbe, A., Goldman, A., Gillis, J. R., Brown, L. R., Toth, R. A., Flaud, J.-M., and Camy-Peyret, C. (1983) AFGL atmospheric line parameters compilation: 1982 edition, Appl. Opt. 22:2247.

4. Rothman, L. S., Goldman, A., Gillis, J. R., Gamache, R. R., Pickett, H. M., Poynter, R. L., Husson, N., and Chedin, A. (1983) AFGL trace gas compilation: 1982 version, Appl. Opt. 22:1616.

(0 - 400  $\text{cm}^{-1}$ ) (millimeter and submillimeter) representing predominantly pure rotational transitions, while the higher frequency domain represents for the most part vibration-rotation transitions. The Trace Gas Compilation currently covers the range 0 to 10,000  $\text{cm}^{-1}$ .

Table 1. Molecular Species on Atmospheric Absorption Line Parameter Atlases\*

Molecule	Isotope	Relative Natural Abundance	Molecule	Isotope	Relative Natural Abundance
$\text{H}_2\text{O}$ (1)	161	0.9973	$\text{HNO}_3$ (12)	146	0.9891
	181	0.0020	OH (13)	61	0.9975
	171	0.0004		81	0.0020
	162	0.0003		62	0.00015
$\text{CO}_2$ (2)	626	0.9842	HF (14)	19	0.99985
	636	0.0110	HCl (15)	15	0.7576
	628	0.0039		17	0.2423
	627	0.0008	HBr (16)	19	0.5068
	638	0.000044		11	0.4930
	637	0.000009	HI (17)	17	0.99985
	828	0.0000040	ClO (18)	55	0.7559
$\text{O}_3$ (3)	728	0.000002		76	0.2417
	666	0.9928	OCS (19)	622	0.937
	668	0.0040		624	0.0416
$\text{N}_2\text{O}$ (4)	686	0.0020		632	0.0105
	446	0.9904		822	0.0019
	456	0.0036	$\text{H}_2\text{CO}$ (20)	126	0.9862
	546	0.0036		136	0.0111
	448	0.0020		128	0.0020
CO (5)	447	0.0004	HOCl (21)	165	0.7558
	26	0.9865		167	0.2417
	36	0.011	$\text{N}_2$ (22)	44	0.9923
	28	0.0020		124	0.9852
$\text{CH}_4$ (6)	27	0.0004	$\text{CH}_3\text{Cl}$ (24)	215	0.7490
	211	0.9883		217	0.2395
	311	0.0111	$\text{H}_2\text{O}_2$ (25)	1661	0.9949
$\text{O}_2$ (7)	212	0.00059	$\text{C}_2\text{H}_2$ (26)	1221	0.9776
	66	0.9952	$\text{C}_2\text{H}_6$ (27)	1221	0.9770
	68	0.0040		1111	0.99955
NO (8)	67	0.0008			
	46	0.9940			
	626	0.9454			
$\text{SO}_2$ (9)	646	0.0420			
	646	0.9916			
$\text{NO}_2$ (10)	646	0.9916			
$\text{NH}_3$ (11)	4111	0.9960			
	5111	0.0036			

\* From the CRC Handbook of Chemistry and Physics, 65th Edition (1985).

An effort is currently underway at AFGL to improve and extend the line compilation for weak water vapor bands and ozone in the visible-UV portion of the spectrum because of the special interest in visible and UV lasers.

### 3. MOLECULAR SCATTERING

In addition to molecular absorption, molecular scattering (Rayleigh) must be included as an attenuation mechanism. The molecular scattering coefficient per unit path length,  $k_{SM}(\nu)$ , at wavenumber  $\nu(\text{cm}^{-1})$  is given by

$$k_{SM}(\nu) = \frac{24 \pi^3 \nu^4}{\rho} \left[ \frac{[n'(\nu)]^2 - 1}{[n'(\nu)]^2 + 2} \right]^2 \left( \frac{6 + 3\delta}{6 - 7\delta} \right) \quad (12)$$

where  $\rho$  is the molecular density of the atmosphere (air density),  $n'(\nu)$  is the real part of the refractive index of air and is a function of wavenumber and air density, and  $\delta$  is the depolarization factor for air ( $\sim 0.03$ ).

### 4. AEROSOL ABSORPTION AND SCATTERING

#### 4.1 Aerosol Absorption

The absorption coefficient due to atmospheric aerosols,  $k_{aA}(\nu)$ , per unit path length at the wavenumber  $\nu$  is given by

$$k_{aA}(\nu) = \int_0^\infty Q_a(x, n) \pi r^2 (dN/dr) dr \quad (13)$$

where  $Q_a$  is the Mie absorption efficiency factor (dimensionless),  $r(\text{cm})$  is the aerosol radius,  $x = 2\pi r\nu$  is the size parameter, and  $n(\nu)$  is the complex refractive index of the aerosol particles.  $dN(r)/dr$  (number/cm<sup>3</sup>/cm) is the aerosol size distribution.

#### 4.2 Aerosol Scattering

Similarly, the aerosol scattering coefficient,  $k_{sA}(\nu)$ , is given by

$$k_{sA}(\nu) = \int_0^\infty Q_s(x, n) \pi r^2 (dN/dr) dr \quad (14)$$

where  $Q_s$  is the dimensionless Mie scattering efficiency factor.

### 4.3 Phase Functions

The angular scattering of radiation by atmospheric molecules or aerosols is specified by phase functions which define the differential probability of radiation being scattered in a given direction. For air molecules, the angular distribution of scattered radiation is given by the Rayleigh scattering phase function:

$$P(\theta) = \frac{3}{16\pi} \left( \frac{2}{2+\delta} \right) [(1+\delta) + (1-\delta) \cos^2 \theta] \quad (15)$$

where  $\theta$  is the scattering angle and  $\delta$  is the depolarization factor for scattering from anisotropic molecules, with the normalization condition

$$\int_{4\pi} P(\theta) d\Omega = 1. \quad (16)$$

Phase functions for aerosols require the use of Mie scattering theory<sup>5</sup> for the angular distribution of radiation and they depend on the size, shape, and complex refractive index of the aerosol particles and on the wavenumber of the radiation.

### 5. TOTAL ATTENUATION

In summary, the total extinction per unit path length,  $k(\nu)$ , due to molecular absorption and scattering and aerosol absorption and scattering is given by the sum of the coefficients defined in Eqs. (11), (12), (13), and (14), that is,

$$k(\nu) = k_{aM}(\nu) + k_{sM}(\nu) + k_{aA}(\nu) + k_{sA}(\nu). \quad (17)$$

For an atmospheric path,  $L$ , the optical depth is equal to  $[k(\nu)L]$ , and the monochromatic transmittance,  $\tau(\nu)$ , is defined as

$$\tau(\nu) = e^{-k(\nu)L}. \quad (18)$$

5. van De Hulst, H. C. (1957) Light Scattering by Small Particles, John Wiley & Sons, New York.

A model and computer code FASCODE (Fast Atmospheric Signature Code) has been developed (Reference 6) for the line-by-line calculation of radiance and transmittance due to line absorption and continuum absorption with spectral line information taken from the Absorption Line Compilation tape(s). Contributions to the total attenuation due to molecular scattering and aerosol absorption and scattering have to be derived at present from LOWTRAN calculations. LOWTRAN (Reference 7) is a band model  $20\text{ cm}^{-1}$  low-resolution transmittance code. It is planned for the future to integrate LOWTRAN and FASCODE with a common set of atmospheric models for aerosols.

## 6. NUMERICAL DATA AND MODEL CALCULATIONS

Descriptions of atmospheric models for water vapor and other atmospheric gases and for atmospheric aerosols have been given in various reports. They are contained in the series of LOWTRAN reports, of which, the most recent is LOWTRAN 6, and in the report on air mass calculations for FASCODE (Reference 14). A detailed description and discussion of boundary layer aerosols is presented in Reference 8. A comprehensive discussion of the optical and infrared properties of the atmosphere will be published as one chapter of a revised Air Force Handbook of Geophysics, currently in publication.

The numerical data for the models of atmospheric gases and aerosols are part of the input data to the transmittance codes, and are available as an integral part of these codes on magnetic tape.\*

Numerical data on spectral extinction coefficients for specific laser frequencies have been derived from model calculations and results have been published for  $\text{CO}_2$ , CO, DF, and HF lasers in References 9, 10, and 11; for HF, DF, and Iodine laser lines in References 12 and 13. Since these reports were published, there have been numerous improvements in the molecular line parameter data, the water vapor and carbon dioxide continuum models, the treatment of molecular line shape, and in the aerosol models. The interested reader is encouraged to implement the transmittance codes, used for the results reported here, to perform calculations for specific conditions of interest and take advantage of continuing developments in this field.

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(Due to the large number of references cited above, they will not be listed here. See References, page 19.)

\*The FASCODE and LOWTRAN computer codes and the molecular absorption line parameter compilations are distributed by the National Climatic Data Center (NOAA), Federal Building, Asheville, NC 28801.

Figures 2 through 7 present the atmospheric transmission properties in the 10.6  $\mu\text{m}$  ( $\text{CO}_2$ ), 3.8  $\mu\text{m}$  (DF), 2.9  $\mu\text{m}$  (HF), 1.3  $\mu\text{m}$  (Iodine), 1.06  $\mu\text{m}$  (Nd-Yag), and visible (Excimer) laser regions. As shown in Table 2, these lasers have high power capability in atmospheric (partially) transparent regions.

Figures 2 and 6 give the transmittance for a 10 km slant path from the surface to 1 km altitude. They give separately the transmittances due to line and continuum absorption, for three different aerosol models, two rain models, and a 100-m thick fog. Multiplication of these individual transmittances together will give total path transmittance. The slant path geometry for these laser transmittance calculations describes a low atmosphere, tactical application. One can see that rain and fog, and aerosols are the dominating attenuators at the wavelengths shown except at several narrow molecular absorption line peaks, especially in the 10.6  $\mu\text{m}$  region.

Figures 3, 4a, 4b, 5, and 7 give ground to space path transmittances for HF, Iodine, and Excimer laser wavelength regions. These plots show only the molecular absorption. Figure 4a gives the transmittance ground to space, Figure 4b from 5 km altitude to space. The difference in transmittance is mostly due to low altitude water vapor absorption. Figure 7 for the visible region shows the transmittances for a ground to space path for ozone, molecular Rayleigh scattering, aerosol extinction, and water vapor and oxygen in the very near IR. As mentioned earlier (Section 2), there are weak absorption bands in the visible from  $\text{H}_2\text{O}$ ,  $\text{O}_3$ , and  $\text{NO}_2$  which are not yet sufficiently characterized to be included in this figure.



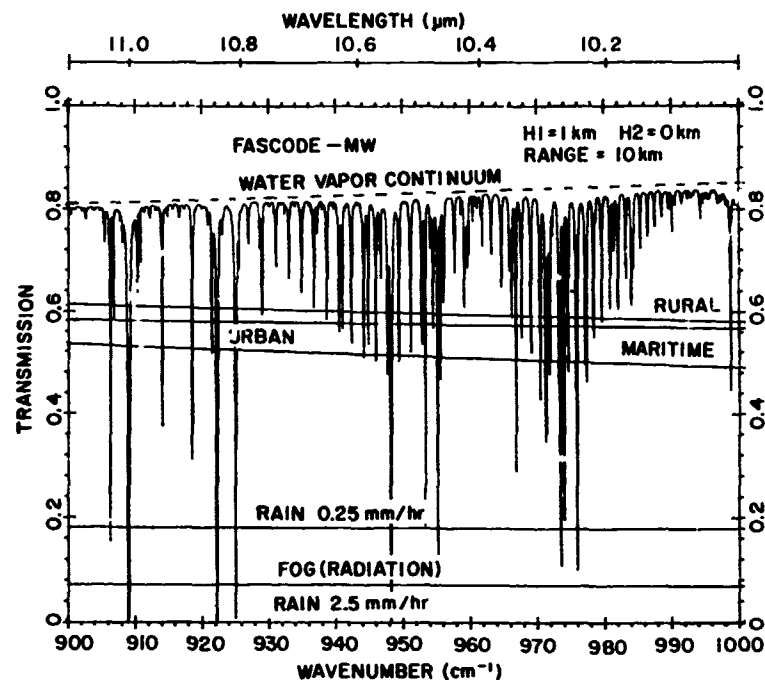


Figure 2. Atmospheric Transmission in the 10.6  $\mu\text{m}$  Spectral Region for a 10 km Slant Path From the Surface to 1 km for the Midlatitude Winter Atmosphere. Attenuation from the total molecular, continuum absorption, aerosols, fog, and rain as indicated; 9400 to 9500  $\text{cm}^{-1}$ . Aerosol concentration corresponds to 10 km visibility; the fog is a 100-m thick layer with 450 m visibility. Transmittance for any combination of the individual attenuation factors would be the product of the total molecular and the aerosol, rain or fog transmission values

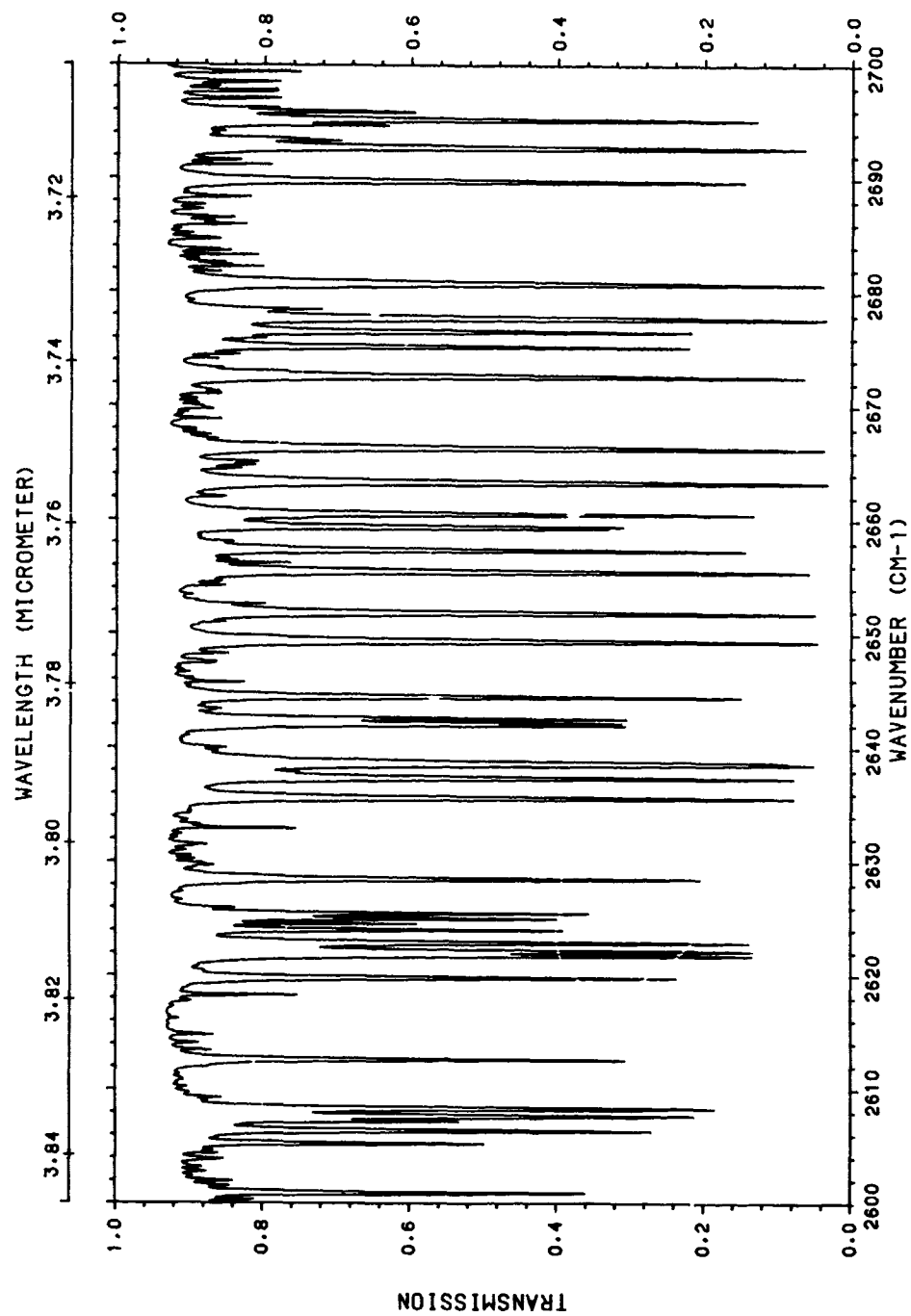


Figure 3. Atmospheric Transmittance for a Vertical Path in the  $3.8 \mu\text{m}$  ( $2600$  to  $2700 \text{ cm}^{-1}$ ) Region Due to Molecular Absorption, Ground to Space

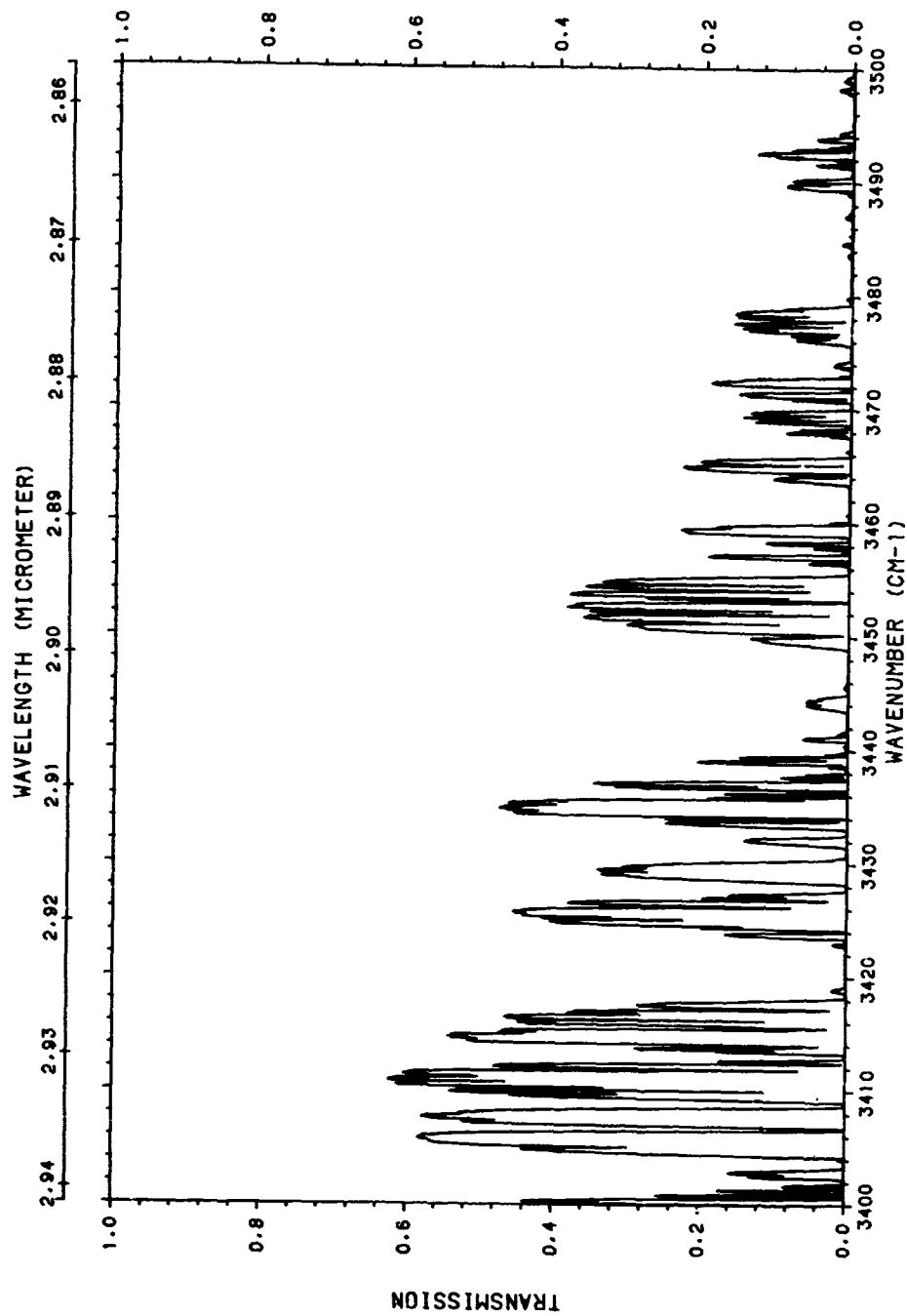


Figure 4a. Atmospheric Transmittance for a Vertical Path in the 2.9  $\mu\text{m}$  ( $3400$  to  $3500\text{ cm}^{-1}$ ) Region Due to Molecular Absorption, Ground to Space

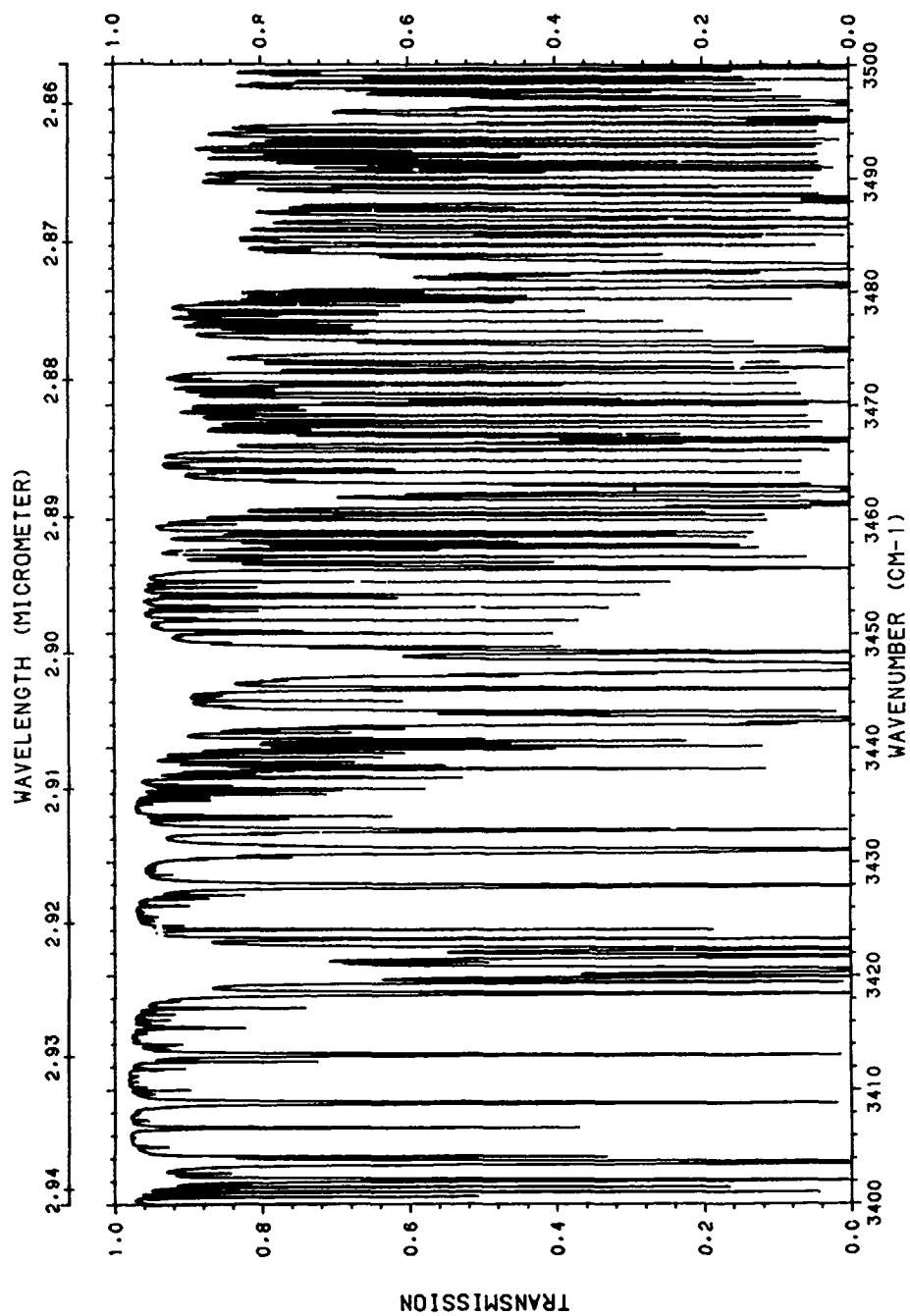


Figure 4b. Atmospheric Transmittance for a Vertical Path in the 2.9  $\mu\text{m}$  (3400 to 3500  $\text{cm}^{-1}$ ) Region Due to Molecular Absorption, 5 km Altitude to Space

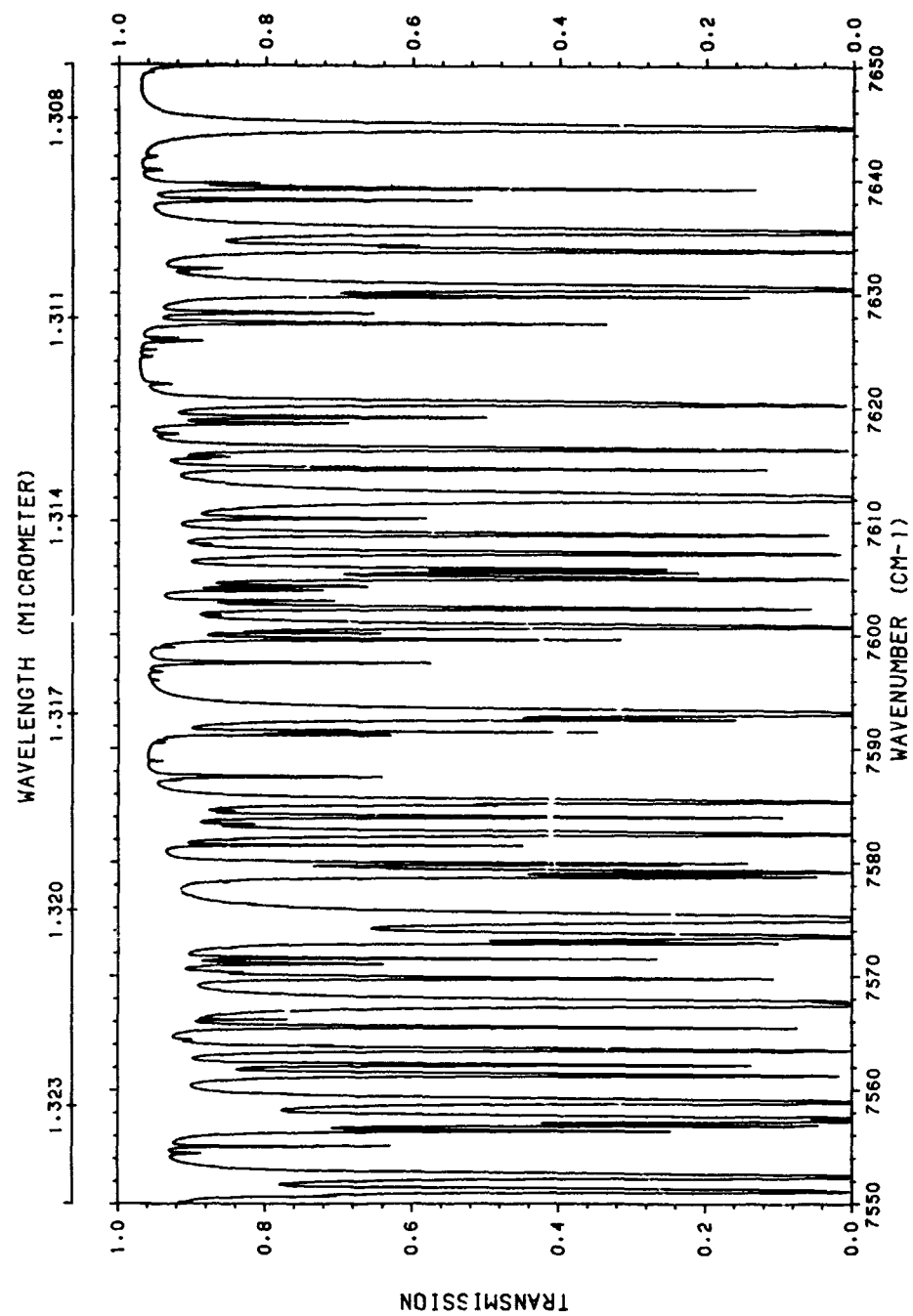


Figure 5. Atmospheric Transmittance for a Vertical Path Ground to Space in the 1.3  $\mu\text{m}$  Region (7550 to 7650  $\text{cm}^{-1}$ ) Due to Molecular Absorption

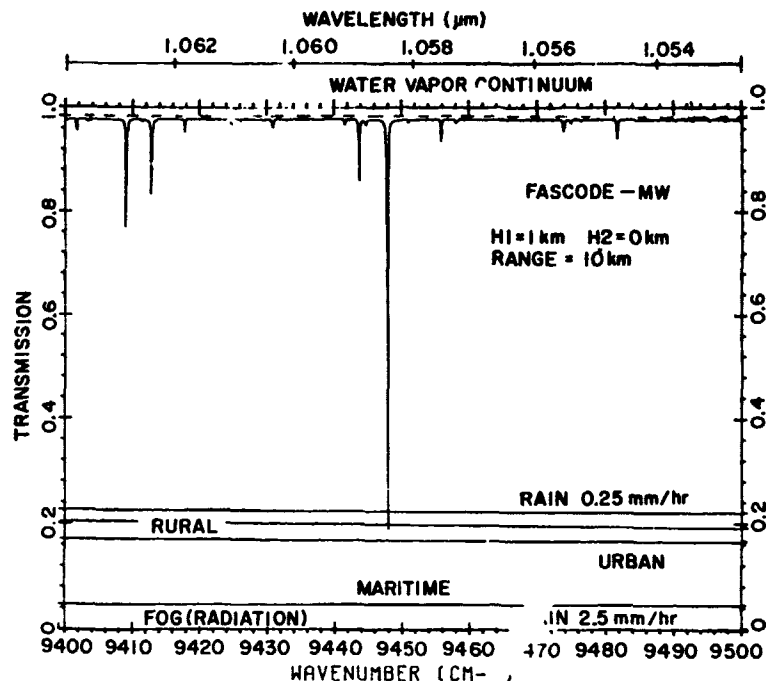


Figure 6. Atmospheric Transmission in the  $10.6 \mu\text{m}$  Spectral Region for a 10 km Slant Path from the Surface to 1 km for the Midlatitude Winter Atmosphere. Attenuation from the total molecular, continuum absorption, aerosols, fog, and rain as indicated; 9400 to 9500  $\text{cm}^{-1}$ . Aerosol concentration corresponds to 10 km visibility; the fog is a 100-m thick layer with 450 m visibility. Transmittance for any combination of the individual attenuation factors would be the product of the total molecular and the aerosol, fog or rain transmission values

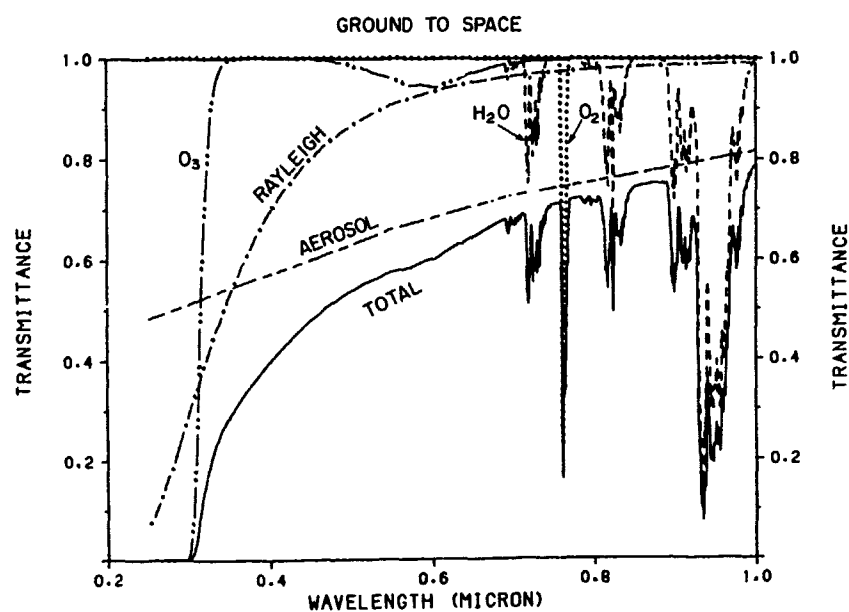


Figure 7. Atmospheric Transmittance for a Vertical Path Ground to Space in the UV-visible Region

Table 2. Most Frequently Referenced Laser Transitions in Atmospheric Partially Transparent Regions

Laser	Identification	Wavenumber cm <sup>-1</sup>	Wavelength μm
CO <sub>2</sub>	P20	944.19403	10.591
DF	P <sub>2</sub> - 8	2631.065	3.801
HF	P <sub>2</sub> - 7	3483.652	2.871
	P <sub>2</sub> - 8	3434.995	2.911
Nd-Yag			1.06
Iodine	3 - 4	7603.3	1.315
Excimers		Tunable	Visible

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